



Article

Advancements in Conductive Cotton Thread-Based Graphene: A New Generation of Flexible, Lightweight, and Cost-Effective Electronic Applications

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Abstract: Conductive threads have emerged as a highly promising platform for the advancement of smart textiles, enabling the integration of conductivity into fabric materials. In this study, we present a novel approach to fabricate highly flexible graphene-based smart threads, which exhibit exceptional electrical properties. Four distinct types of smart threads were meticulously prepared by drop-casting graphene dispersions onto cotton threads, utilizing various solvents. The influence of annealing temperature and the quantity of dispersed graphene on the electrical conductivity of the threads was systematically investigated. Our findings reveal that the electrical conductivity of the threads is significantly influenced by the type of solvent and the annealing temperature, while exhibiting an increasing trend with higher amounts of dispersed graphene. Remarkably, we achieved a maximum electrical conductivity of $2505.68 \text{ S cm}^{-1}$ for a thread prepared with 6 mL of graphene dispersed in ethanol, annealed at a temperature of 78°C . Furthermore, the fabricated smart threads were successfully employed as replacements for electric cables in a mobile charger and a computer mouse, demonstrating their high efficiency. This work represents a significant advancement in the development of a new generation of smart textiles, offering a simple, cost-effective, and environmentally friendly fabrication method for the production of smart threads.

Keywords: graphene; smart material; eco-friendly; conductive; nanomaterial; technology



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1. Introduction

Conductive threads have emerged as a promising material for various applications in the field of electronics and textiles. These threads possess high electrical conductivity, making them suitable for integration into electronic textiles, wearable devices, and smart fabrics. The seamless integration of conductive threads into clothing and accessories enables the development of interactive and functional garments [1–8]. For instance, health monitoring sensors embedded in clothing can provide real-time physiological data, facilitating personalized healthcare and wellness monitoring. Additionally, conductive threads can be utilized to create touch-sensitive interfaces on wearable devices, enhancing user interaction and control. The unique properties of conductive threads offer a novel approach to merging technology with everyday textiles, enabling the realization of innovative applications that enhance user experience and enable the seamless integration of electronics into our daily lives [9–13].

There are different types of smart materials used for the production of conductive threads, including conventional options such as carbon nanomaterial [14,15], metal alloys [16], and polymers. However, the integration of advanced functionalities into cotton threads has remained a challenge.

In recent years, graphene, a two-dimensional material composed of a single layer of carbon atoms, has gained significant attention in the field of conductive thread manufacturing [17–20]. Its exceptional electrical, mechanical, and thermal properties make it an ideal

candidate for enhancing the performance of conductive threads. By incorporating graphene into the thread's structure, researchers have been able to achieve even higher levels of electrical conductivity, enabling more efficient transmission of electrical signals [21–27]. Furthermore, graphene's unique properties allow for the production of conductive threads that are lightweight, durable, and resistant to environmental factors such as moisture and temperature fluctuations. These advancements in graphene-based conductive threads hold great promise for the development of next-generation electronic textiles and wearable devices, opening up new possibilities for applications in healthcare, sports, fashion, and beyond.

Several studies have explored the integration of graphene into textile materials to create graphene-infused threads with enhanced properties. In a recent study [28] conducted by Li et al. (2023), graphene-coated threads with improved thermoelectric properties were developed using a 3D extrusion method. The resulting thermoelectric threads demonstrated excellent stability over 1000 cycles, indicating their potential suitability for strain sensor applications in thermally regulated textiles and wearable devices. These findings highlight the successful integration of graphene into threads, leading to enhanced thermoelectric performance and potential applications in the field of wearable technology. In their study [29], Tan et al. (2022) investigated the application of reduced graphene oxide (rGO) as a coating material for cotton threads. The researchers found that integrating rGO onto cotton threads resulted in increased electrical conductivity. When these threads were stitched into a piezoresistive sensor, they also showed improved sensitivity and cyclability. These results highlight the potential of rGO-coated cotton threads for the development of high-performance piezoresistive sensors. In a study by Zhong et al. (2013), a composite material of graphene and double-walled carbon nanotubes was successfully integrated into threads [9]. This integration was achieved by depositing the composite material on the thread surface using chemical vapor deposition (CVD). The resulting threads exhibited enhanced electrical conductivity of 10^5 S m^{-1} and improved mechanical strength of 300 MPa. Despite these advancements, challenges still exist in the fabrication of graphene-infused threads. For instance, achieving a uniform and stable dispersion of graphene within the textile matrix remains a key challenge. Various techniques, such as solution blending, electrospinning, and dip coating, have been explored to address this issue. Additionally, the scalability and cost-effectiveness of the fabrication processes need to be improved to enable large-scale production of graphene-infused threads.

In this study, we present the fabrication of highly conductive cotton threads through the dispersion of graphene, aiming to investigate the influence of different solvents on the electrical properties of the resulting conductive cotton threads. Four solvents, namely, deionized water, dimethylformamide, dimethyl sulfoxide, and ethanol, were utilized for the dispersion process. These solvents were selected due to their diverse properties and compatibility with graphene dispersion. Each of these solvents offers unique advantages in terms of dispersing graphene and affecting the resulting electrical properties of the composite threads. By using these solvents, we sought to systematically investigate their influence on the electrical conductivity of the composite threads when graphene is dispersed in them. This comprehensive analysis provides valuable insights into the suitability of different solvents for specific applications, as well as the ability to tailor the electrical performance of the fabricated conductive cotton threads. Additionally, we explore the impact of annealing temperature on the electrical conductivity of the conductive filaments, employing two annealing temperatures: one near the boiling point and another far from the boiling point of the solvent. Our investigation focuses on the potential replacement of copper wire with conductive graphene threads in mobile chargers and computer mice. Through rigorous experimentation and analysis, we assess the efficiency and performance of these conductive threads in these applications. The results obtained demonstrate the high efficiency and functionality of the conductive threads, highlighting their promising potential as a viable alternative to traditional copper wire. By elucidating the relationship between solvent selection, annealing temperature, and the electrical properties of the conductive cotton

threads, this study provides valuable insights into the optimization of fabrication processes for graphene-based conductive materials. Furthermore, our findings contribute to the advancement of sustainable and efficient technologies in the field of electronic devices. While it is true that graphene-coated threads have been extensively studied in various electronic applications, our work presents a novel and simplified approach to fabricating conductive cotton threads with dispersed graphene. The significance of our study lies in the simplicity and environmental friendliness of the fabrication process. We employed a straightforward method utilizing four different solvents, which not only simplifies the production process but also minimizes chemical waste generation. This approach is in line with the growing interest in sustainable and eco-friendly manufacturing practices, which is particularly important in today's environmentally conscious society. Furthermore, by systematically investigating the influence of annealing temperature and graphene quantity on the electrical conductivity of the composite threads, we provide valuable insights into the optimization of these materials for specific electronic applications. Our work aims to contribute to the field by offering an efficient and environmentally responsible method for producing conductive cotton threads with graphene, which can have applications in various new-generation electronic devices. We believe that the simplicity of our method and its potential environmental benefits make it a valuable addition to the existing body of research on conductive cotton/graphene composites.

2. Experimental Section

2.1. Materials

Cotton threads were obtained from FPC created technical textiles, Saudi Arabia, and used as the substrate. Graphene powder, purchased from Sigma Aldrich, Gillingham, UK, was utilized as the conductive material in this study. Dimethylformamide (DMF), dimethyl sulfoxide (DMSO), and ethanol, purchased from Sigma Aldrich, were employed as solvents without any additional purification.

2.2. Preparation of Graphene Dispersion

Four distinct graphene dispersion solutions were prepared using the following procedure. For the first solution, 600 mg of graphene powder was accurately weighed and mixed with 3 mL of deionized water. The mixture was then subjected to sonication at a temperature of 24 °C for a duration of 20 min, ensuring the formation of a homogeneous solution. The same procedure was repeated for the remaining solutions, with the only variation being the substitution of the organic solvents dimethylformamide (DMF), dimethyl sulfoxide (DMSO), and ethanol in place of deionized water.

2.3. Fabrication of Conductive Cotton Threads

The cotton threads used in the experiment were first washed with deionized water and dried. To prepare the conductive threads (see Figure 1), a dry thread measuring approximately 10 cm in length was immersed in deionized water for 10 min. Following the immersion, 1 mL of the graphene dispersion solution was carefully dripped onto the thread to ensure even distribution and adherence of the solution. The treated threads were then stored at a temperature of 24 °C for 10 min to allow for proper absorption and distribution of the graphene dispersion. Subsequently, the threads were dried in an oven at a specific annealing temperature, which was chosen to be close to or far from the boiling point of the solvent used in the graphene dispersion solution. Table 1 mentions the annealing temperatures used. To increase the amount of graphene on the thread, the immersion process was repeated multiple times. It is worth noting that the experiment was conducted using two different annealing temperatures, and a total of 60 conductive cotton threads were produced. Table 1 provides the boiling points of the solvents used, the annealing temperatures, and the number of conductive cotton threads produced.

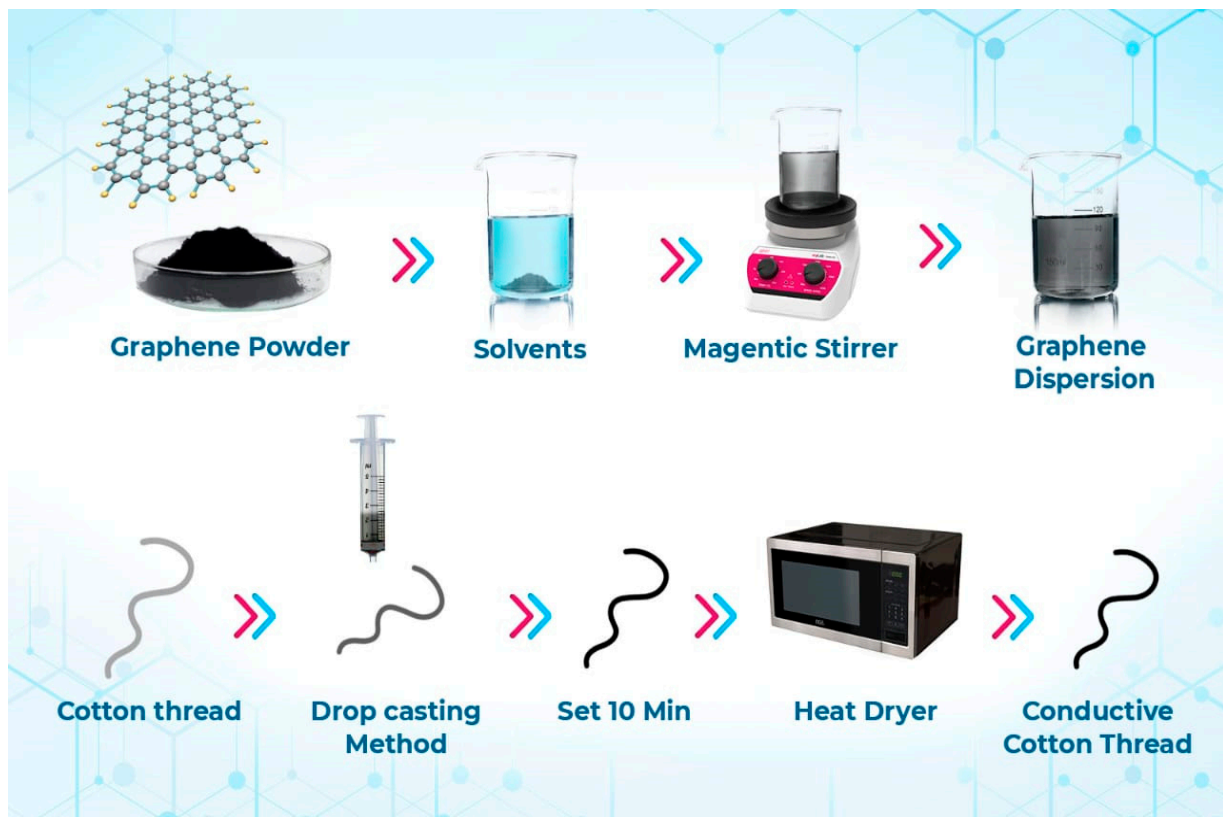


Figure 1. Schematic diagram of the process for the production of conductive graphene–cotton threads using the method of “drop-casting and drying”.

Table 1. The boiling points of the solvents used, the temperatures at which the samples were dried, and the number of conductive cotton fabrics produced.

Graphene Dispersion in	Boiling Point (°C)	T ₁ (°C)	Number of Samples	T ₂ (°C)	Number of Samples
DI	100	100	8	85	8
DMSO	189	180	8	100	8
DMF	153	153	7	100	7
Ethanol	78.4	78	7	60	7

2.4. Characterization and Measurements

The surface structure of the untreated and treated cotton threads was studied by scanning electron microscopy (SEM) (JSM-7610F Schottky Field Emission Scanning Electron Microscope, DirectIndustry, 17 avenue André Roussin, Marseille, France) and elemental structure analysis by energy dispersive X-ray analysis (EDS). A PerkinElmer FTIR analyzer was used to investigate the chemical composition of the conductive threads under ambient conditions in the wavenumber range 4000–500 cm^{−1}. The TG/DTA Model DTG-60 simultaneous instrument was used for thermogravimetric analysis (TGA) and differential thermal analysis (DTA) under a dynamic nitrogen atmosphere with a heating rate of 50 °C/min from 30 °C to 900 °C. The homemade four-line probe technique was used to study the electrical properties of the conductive filaments, measuring the current and differential voltage with a Peak Toch 3340 DMM multimeter. The electrical resistance (*R*) was then calculated from the *I*–*V* curve at 24 °C and 65% relative humidity, and then the electrical conductivity was calculated using the relationship $\sigma = l/RA$, where *l* is the filament length (10 cm) and *A* is the cross-sectional area (0.0314 cm²).

3. Results and Discussion

3.1. SEM Analysis

Scanning electron microscopy (SEM) was employed to investigate the surface morphology of the conductive cotton threads coated with graphene. Figure 2 presents the SEM images, where Figure 2A represents the untreated cotton thread, while Figure 2B through 2D depict the treated cotton thread at varying concentrations of graphene dispersed in deionized water. Figure 2A reveals elongated and smooth fibers with loose packing, resulting in a small, fluffy, and porous structure. The average diameter of the fibers measures 6.40 μm . In Figure 2B, the presence of graphene coating on some fibers is evident, and as the concentration of graphene increases, a greater number of fibers become coated, leading to the filling of inter-fiber spaces, as observed in Figure 2C,D. The enhanced brightness of Figure 2D, in comparison to Figure 2A, indicates the successful transformation of the cotton threads into electrically conductive entities.

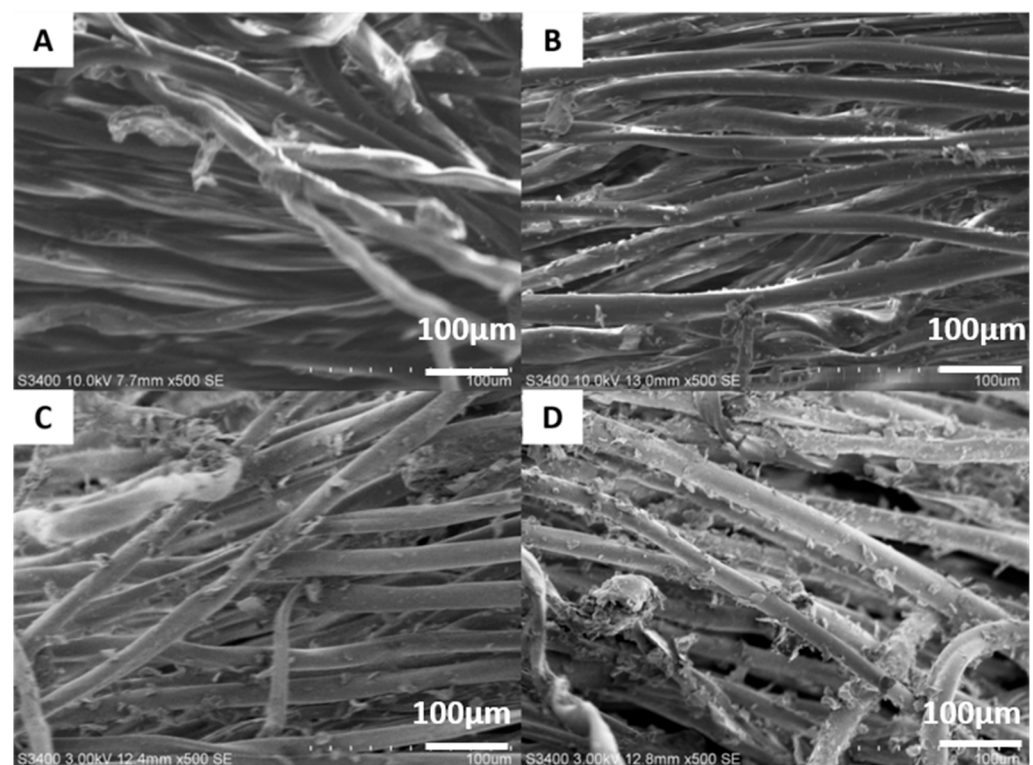


Figure 2. SEM images of untreated cotton thread (A) and treated cotton thread with low, medium, and high concentrations of graphene dispersed in deionized water (B–D). The SEM images show a typical smooth fiber whose morphology changes significantly after graphene coating on the conductive cotton thread surface.

3.2. EDS Analysis

Figure 3 presents the energy dispersive X-ray spectroscopy (EDS) analysis results obtained from the untreated cotton thread and cotton threads treated with varying concentrations of graphene. In the EDS analysis of the untreated cotton thread (Figure 3A), the presence of carbon and oxygen is observed, which can be attributed to the cellulose structure inherent in cotton fibers [30,31]. Additionally, the presence of silica is detected, as expected in commercial cotton threads, as silica is commonly incorporated in industrial applications to enhance thread strength, durability, and resistance to heat, chemicals, and abrasion [32]. The EDS spectra of the graphene-treated cotton thread (Figure 3B,C) exhibit the presence of carbon, oxygen, and silica, with an increasing weight fraction of carbon. This observation provides compelling evidence for the successful deposit of graphene on the cotton surface. The incorporation of graphene onto the cotton thread surface offers the

potential for enhanced mechanical properties and improved performance characteristics, owing to graphene's exceptional strength, electrical conductivity, and chemical stability. These findings underscore the promising prospects of utilizing graphene as a functional coating material for cotton threads, opening up avenues for the development of advanced textiles with enhanced properties and performance.

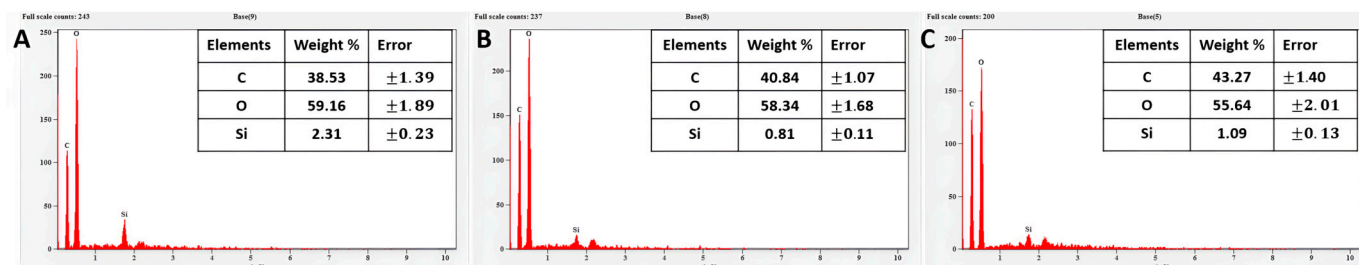


Figure 3. EDS spectra of untreated cotton thread (A) and graphene-treated cotton thread at low and high graphene concentrations (B) and (C), respectively.

3.3. FTIR Analysis

FTIR analysis of graphene powder and untreated and treated cotton threads with graphene dispersion was studied in the wavenumber range from 400 to 4000 cm^{-1} , as Figure 4 shows. The main purpose of FTIR analysis of the treated cotton thread is to investigate the effects of solvents, graphene concentration, and annealing temperature (see Table 2). In the FTIR spectra of graphene powder, the presence of strong characteristic peaks at 672 cm^{-1} and 950 cm^{-1} corresponds to the bending vibrations of the C-H and C-O bonds, respectively. These peaks offer evidence of the chemical composition and functional groups present in the graphene material. For the untreated cotton thread, the FTIR spectra revealed characteristic peaks at 2900 cm^{-1} , indicative of the C-H stretching vibrations within the β -glucose unit of cellulose, and at 1750 cm^{-1} and 1019 cm^{-1} , associated with the C=O stretching of carboxylic acid and C-O stretching, respectively. Additionally, the bands observed at 1800 cm^{-1} and 2250 cm^{-1} represent C=C vibrational modes. These findings provide valuable information regarding the chemical constituents and structural attributes of the untreated cotton thread [30]. The indication of the FT-IR spectrum lies in its ability to detect and differentiate these characteristic peaks, which can be attributed to specific chemical bonds and molecular groups. The FTIR results of graphene powder and the untreated cotton thread are in good agreement with the literature [33,34]. Moreover, through the FTIR analysis of the treated cotton threads, we aimed to investigate the impact of various factors, including solvents, graphene concentration, and annealing temperature, on the FTIR spectra. The absence or alterations of certain peaks under different experimental conditions offer insights into the interactions and modifications occurring within the treated cotton threads.

Table 2. The details of the conductive threads used in FTIR analysis.

Solvents	Boiling Point 1	Concentration	Boiling Point 2	Concentration
DI	100 °C	11 wt. %	85 °C	21 wt. %
		35 wt. %		33 wt. %
		45 wt. %		69 wt. %
DMSO	180 °C	21 wt. %	100 °C	11 wt. %
		26 wt. %		50 wt. %
		41 wt. %		66 wt. %

Table 2. Cont.

Solvents	Boiling Point 1	Concentration	Boiling Point 2	Concentration
DMF	153 °C	13 wt.%	100 °C	19 wt.%
		35 wt.%		49 wt.%
		43 wt.%		55 wt.%
Ethanol	78 °C	19 wt.%	60 °C	30 wt.%
		40 wt.%		40 wt.%
		55 wt.%		58 wt.%

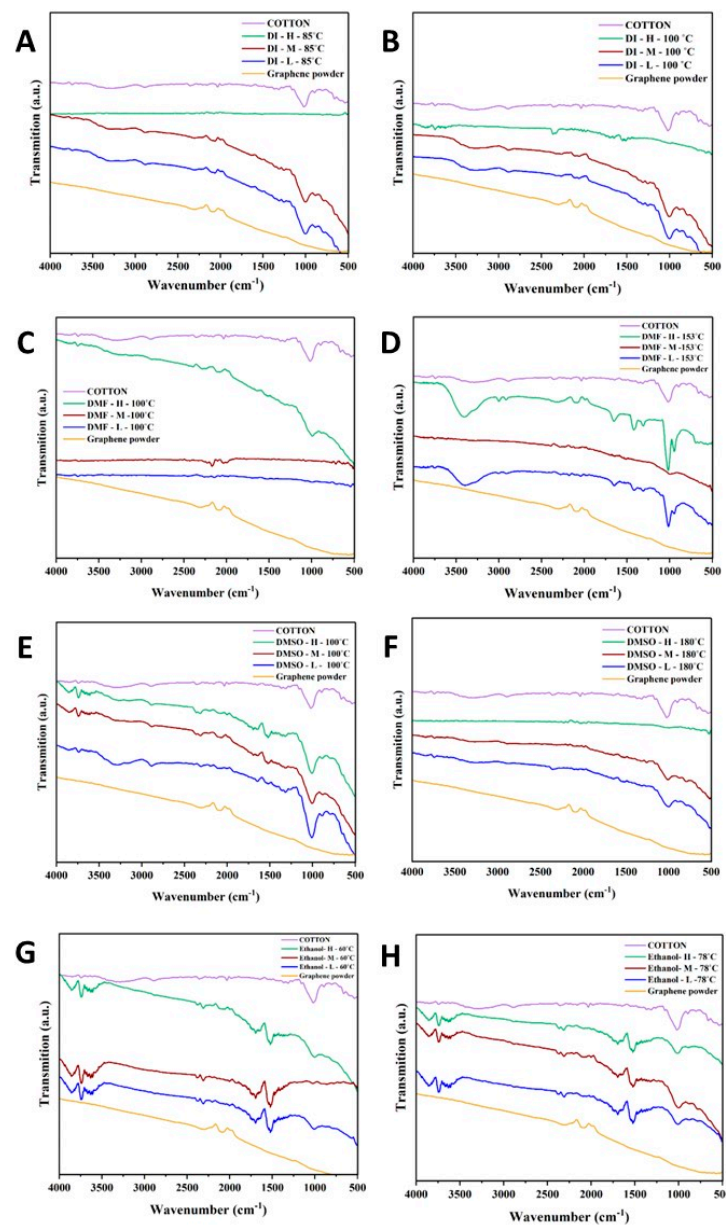


Figure 4. FTIR spectra of the conductive threads at different amounts of graphene dispersed in DI (A,B), DMF (C,D), DMSO (E,F), and ethanol (G,H) at two annealing temperatures. The purple, green, brown, blue, and yellow curves in each figure show the FTIR spectra of the untreated cotton thread and the cotton threads treated with high, medium, and low amounts of graphene dispersion.

3.4. Thermal Analysis

The TGA and DTA analyses of the graphene powder and untreated and treated cotton threads were examined to determine the thermal stabilities and degradation profiles (see Figure 5). Table 3 lists the thermal onset and endset temperatures and thermal stability of the three samples. TGA analysis of graphene powder (red curve) shows that graphene has high thermal stability with low mass loss in the temperature range of 30 to 615 °C and then degrades by combustion of the carbon skeleton. The broad endothermic peak observed in the DTA analysis of the graphene powder (Figure 5B) is followed by an exothermic peak at 700 °C. TGA analysis of the untreated cotton thread (purple curve) showed that the sample is stable in the temperature range from 30 to 366 °C with a mass loss of 21.17%. After that, the sample started to decompose and reached the maximum decomposition at 433.45 °C, indicating an endothermic peak, as shown in the DTA analysis with a mass loss of 73.96%. TGA analysis of the treated cotton thread was performed for the cotton thread treated with graphene dispersed in DI. The TGA curve (green curve) showed that the treated thread is stable in the range from 30 to 371.73 °C with a mass loss of 17.61%, which means that the thermal stability of the thread is increased by 4.45% and with less mass loss by the addition of graphene nanoparticles compared to the untreated thread. The decomposition process started at 370.76 °C and reached the maximum decomposition at 424.31 °C, indicating an endothermic peak, as shown in the DTA analysis with a mass loss of 77.07%.

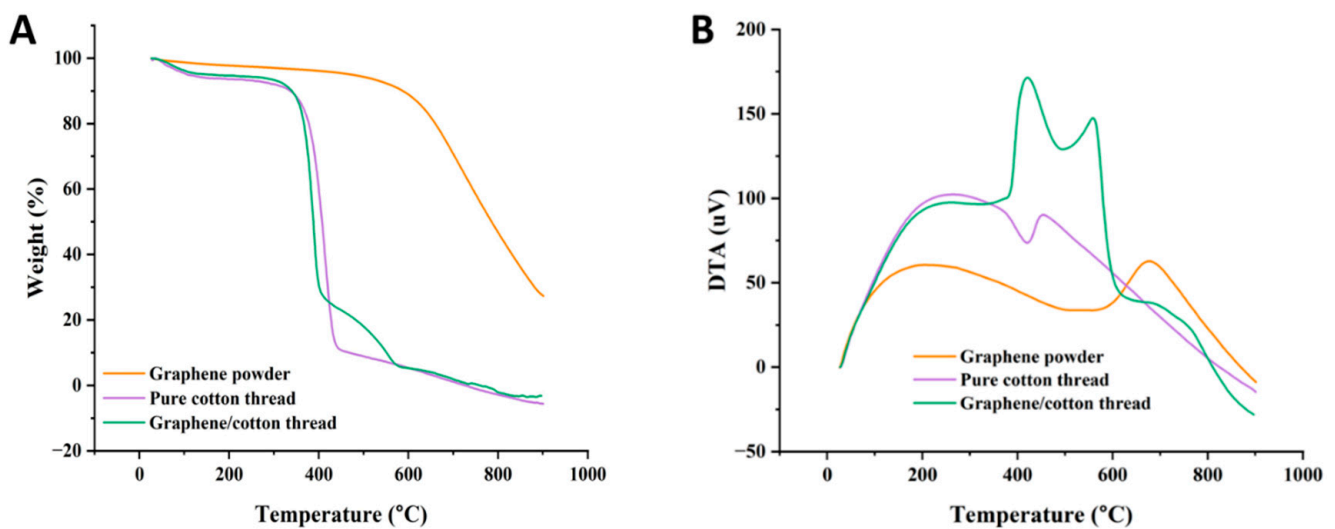


Figure 5. TGA analysis (A) and DTA analysis (B) of pure cotton graphene powder and conductive threads.

Table 3. TGA analysis of graphene powder and untreated and treated cotton thread.

Sample	Onset Temperature (°C)	Endset Temperature (°C)	Weight Loss (%)	Stability Range °C
Graphene powder	510.83	616.83	7.6	30–615
Untreated thread	393.19	433.45	73.9	30–330
Treated thread	386	442.30	77	30–345

The thermal analysis revealed that the graphene powder (red curve) exhibits high thermal stability with minimal mass loss between 30 to 615 °C, degrading thereafter due to the combustion of its carbon skeleton; its DTA analysis showed a broad endothermic peak followed by an exothermic one at 700 °C. In contrast, the untreated cotton thread remains stable from 30 to 366 °C, experiencing a 21.17% mass loss, and begins decomposing post this range, reaching its peak decomposition at 433.45 °C with a 73.96% mass loss. When treated with graphene, the cotton thread's stability improves, evident from its 17.61% mass

loss between 30 to 371.73 °C. This treatment enhances its thermal stability by 4.45% and reduces mass loss compared to the untreated version. Decomposition for the treated thread initiates at 370.76 °C, peaking at 424.31 °C with a 77.07% mass loss.

3.5. Electrical Conductivity Measurements

In this experiment, the effect of three independent variables, namely, the amount of graphene dispersion dispersed in different solvents, the annealing temperature, and temperature on the electrical conductivity, which is the dependent variable, is investigated. The electrical conductivity was calculated for the four solvents at graphene dispersion amounts of 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, and 6.0 mL. This calculation was performed at two annealing temperatures: far from the solvent boiling point and close to it. It is worth noting that the total number of samples in this study was 56 samples.

First, the effect of the amount of graphene dispersion in different solvents on the electrical conductivity of the conductive threads is discussed. As Figure 6 shows, the electrical behavior of all threads is the same at the two annealing temperatures. The conductivity increases with the increasing amount of graphene dispersion in the thread which is due to the increase in graphene concentration in the thread. Figure 6 shows an exponential curve, and with a small amount of graphene dispersion, the value of conductivity is not clear. Moreover, some values of conductivity in this range also overlap. To solve this problem and find a relationship between electrical conductivity (σ) and the amount of graphene dispersion (V), we plotted the natural logarithm of conductivity against the natural logarithm of graphene dispersion (see Figure 7). The experimental data of these curves were fitted to the model:

$$\ln \sigma = \alpha \ln V + \beta \quad (1)$$

where α and β are the fitting parameters (see Table 4) which are calculated using the least square method. All conducting threads give well-fitting straight lines. From Equation (1), we can conclude that σ is proportional to V^α according to the following equation:

$$\sigma = CV^\alpha \quad (2)$$

where $C = e^\beta$ and Table 5 shows its values.

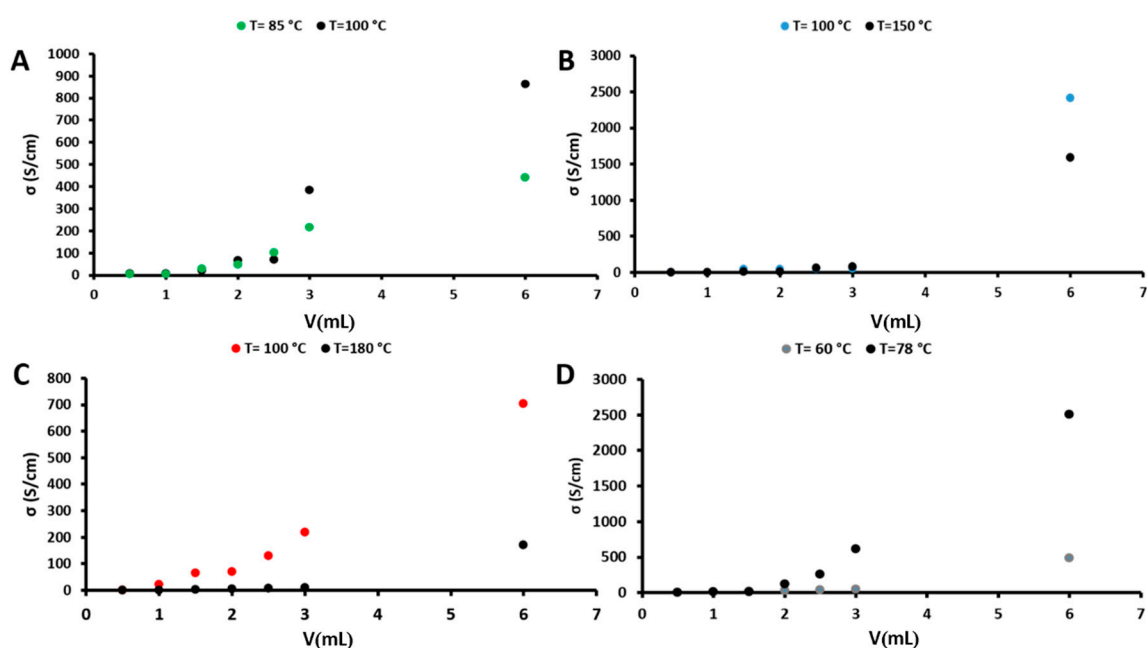


Figure 6. Electrical conductivity of the conductive thread as a function of graphene dispersion in (A) DI, (B) DMF, (C) DMSO, and (D) ethanol at two annealing temperatures.

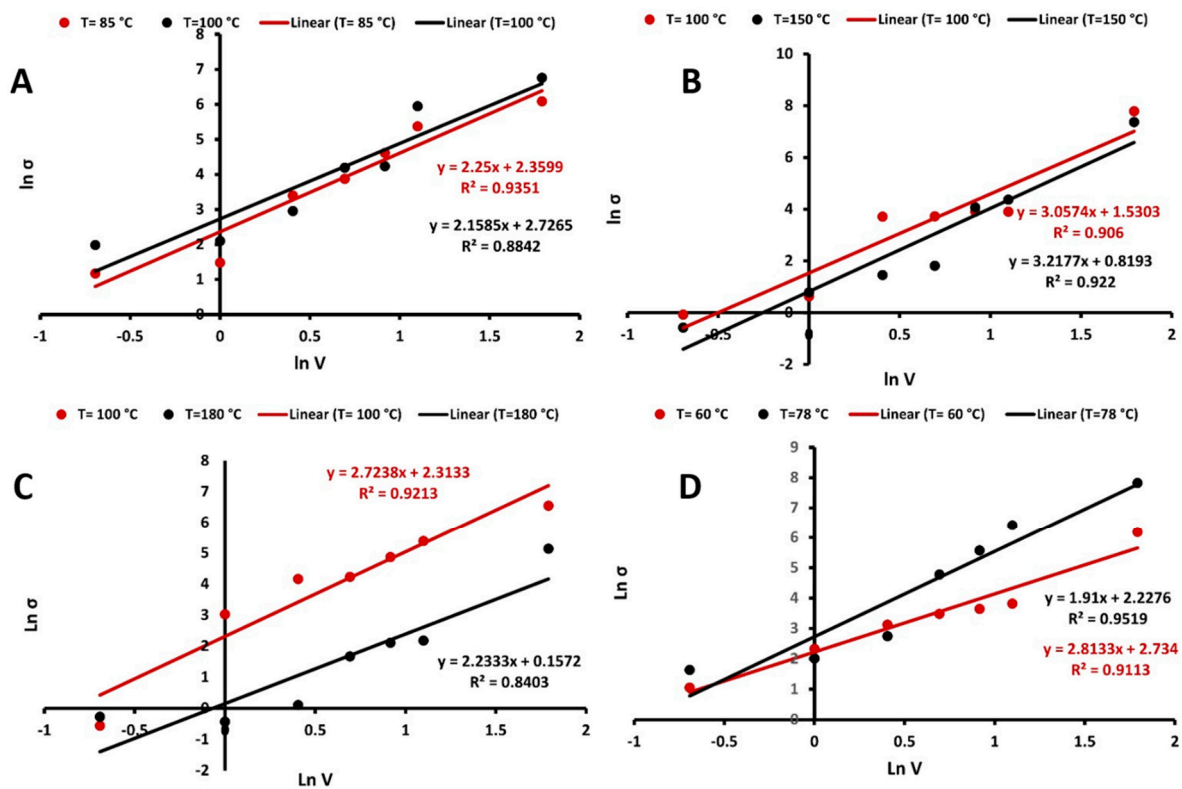


Figure 7. Theoretical analysis of the electrical conductivity of the conductive thread to find the best-fitting straight line describing the relationship between the conductivity and the graphene dispersion in (A) DI, (B) DMF, (C) DMSO, and (D) ethanol at two annealing temperatures.

Table 4. The fitting parameters α , β , C , and R^2 .

Graphene Dispersion in	DI		DMSO		DMF		Ethanol	
	T = 85 °C	T = 100 °C	T = 100 °C	T = 180 °C	T = 100 °C	T = 150 °C	T = 60 °C	T = 78 °C
α	2.25	2.1585	2.7238	2.2333	3.0574	3.2177	2.8133	1.91
β	2.3599	2.7265	2.3133	0.1572	1.5303	0.8193	2.734	2.2276
R^2	0.9351	0.8842	0.9213	0.8403	0.906	0.922	0.9113	0.9519
$C = e^\beta$	10.5898	15.2793	10.1077	1.17022	3.7184	2.2689	15.3943	9.2803

Table 5. Electrical conductivities as a function of graphene dispersion at two annealing temperatures.

V (mL)	Electrical Conductivity (S cm ⁻¹)							
	Graphene Dispersion in DI		Graphene Dispersion in DMSO		Graphene Dispersion in DMF		Graphene Dispersion in Ethanol	
	T = 85 °C	T = 100 °C	T = 100 °C	T = 180 °C	T = 100 °C	T = 150 °C	T = 60 °C	T = 78 °C
0.5	3.21623	7.25249	0.572574	0.764215	0.93232	0.560689	2.863178	5.141029
1	4.390467	8.191767	20.56113	0.651404	1.875015	2.190011	10.26268	7.46254
1.5	29.73311	19.28026	63.98604	1.119801	40.82966	4.28629	22.74795	15.56632
2	48.07115	66.05919	69.08871	5.298671	41.35991	6.153798	32.49708	118.9658
2.5	101.0924	68.78431	130.0945	8.218615	49.65253	58.65034	38.37004	262.2026
3	216.3528	384.256	217.7284	8.88344	49.6525	78.891	45.49591	614.9282
6	441.8304	863.7682	702.717	169.5801	2416.323	1589.178	486.5872	2505.675

Secondly, we want to investigate whether or not the annealing temperature affects the electrical conductivity of the conductive thread. For this purpose, the conductivity was studied as a function of solvent type for a fixed amount of graphene dispersion. As Figure 8 and Table 5 show, the conductivity of the conductive thread depends strongly on the annealing temperature. For the conductive thread prepared with graphene dispersed in DMSO, the conductivity of the thread prepared at a temperature far from the boiling point ($T = 100\text{ }^{\circ}\text{C}$) is higher than the conductivity of the thread prepared at a temperature close to the boiling point ($T = 180\text{ }^{\circ}\text{C}$) for both the low and high graphene dispersion samples. The maximum conductivity was obtained as 702.717 S cm^{-1} at an annealing temperature of $100\text{ }^{\circ}\text{C}$ and 6 mL of graphene dispersion. In contrast, the conductivity of the conductive thread prepared with graphene dispersed in DI at an annealing temperature far from the boiling point ($T = 85\text{ }^{\circ}\text{C}$) is lower than the conductivity at an annealing temperature near the boiling point ($T = 100\text{ }^{\circ}\text{C}$). The maximum conductivity was determined to be 863.768 S cm^{-1} at an annealing temperature of $100\text{ }^{\circ}\text{C}$ and 6 mL of graphene dispersion. For the conductive thread prepared with graphene dispersed in DMF, the conductivity of the conductive thread at a graphene dispersion amount of 1 mL and 3 mL is approximately the same at both annealing temperatures. In contrast, the conductivity of the conductive thread at an annealing temperature far from the boiling point ($T = 100\text{ }^{\circ}\text{C}$) is higher than the conductivity at an annealing temperature near the boiling point ($T = 150\text{ }^{\circ}\text{C}$) for the conductive thread prepared with 2 mL and 6 mL of graphene dispersion, and the maximum conductivity of $2416.323\text{ S cm}^{-1}$ was obtained at an annealing temperature of $100\text{ }^{\circ}\text{C}$ for 6 mL of graphene dispersion. For the conductive thread prepared with graphene dispersed in ethanol, the conductivity of the thread prepared at an annealing temperature near the boiling point is higher than the conductivity of the thread prepared at an annealing temperature far from the boiling point for all samples except the thread prepared with a small amount of graphene dispersion. The maximum conductivity of $2505.675\text{ S cm}^{-1}$ was obtained at an annealing temperature of $78\text{ }^{\circ}\text{C}$ and a high amount of graphene dispersion.

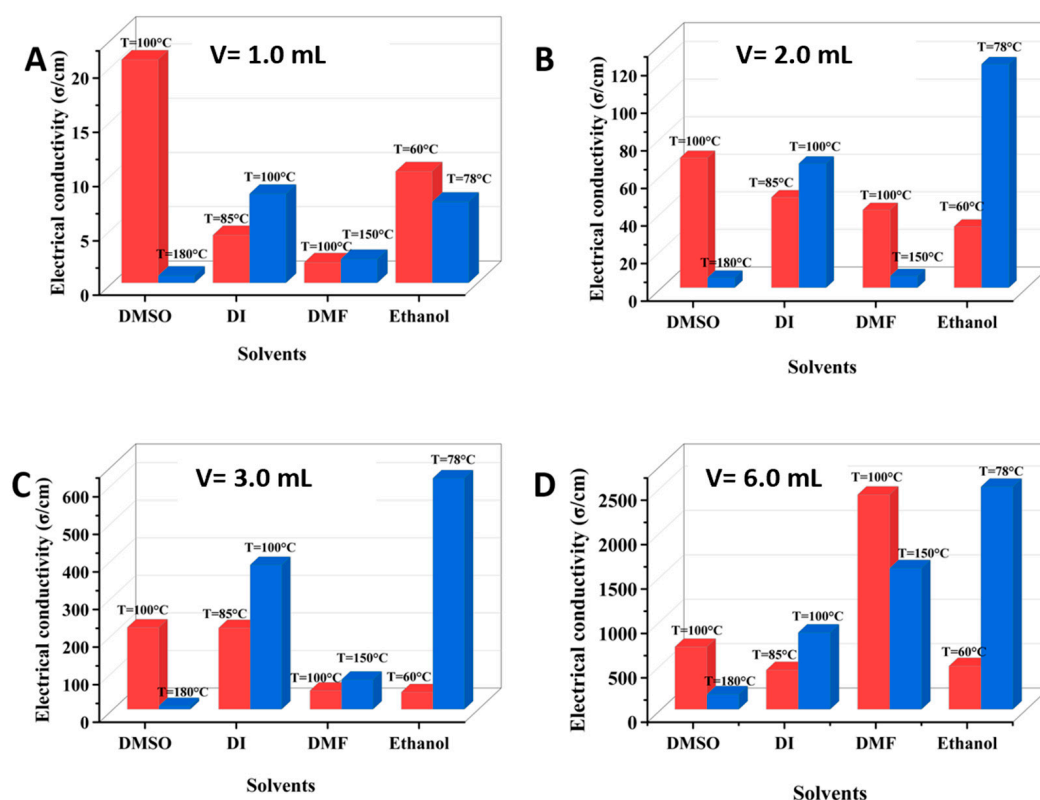


Figure 8. Electrical conductivity of the conductive thread as a function of solvent type (A) DI, (B) DMF, (C) DMSO, and (D) ethanol at two annealing temperatures.

The influence of the amount of graphene dispersion and annealing temperature on the electrical conductivity of the thread can be explained through the complex interplay of several factors at the nanoscale. Firstly, the amount of graphene dispersion directly impacts the concentration of graphene within the composite material. Graphene, being an excellent conductor of electricity, introduces more conductive pathways within the material. This increase in the number of conductive paths enhances the overall electrical conductivity of the thread. Secondly, the annealing temperature plays a crucial role in the structural arrangement of graphene within the thread. At elevated temperatures, graphene sheets tend to align more uniformly, reducing defects and discontinuities in the conductive pathways. This alignment enhances the charge transport efficiency, further contributing to increased electrical conductivity. Additionally, annealing can remove residual solvents or binders, which may hinder electron movement within the composite. By eliminating these impediments, annealing promotes better electrical connectivity between graphene sheets and the surrounding matrix. In summary, the amount of graphene dispersion and annealing temperature affect the electrical conductivity of the thread by influencing the concentration of graphene, the structural arrangement of graphene sheets, and the removal of hindrances to electron movement. These factors collectively contribute to the observed variations in electrical conductivity, making them critical parameters to optimize for tailored electrical performance in the composite material.

Finally, we have successfully fabricated highly conductive cotton threads through a dip-and-dry method using graphene as the substrate material. Our research has yielded remarkable results, with the cotton threads exhibiting an impressive maximum electrical conductivity of $2505.675 \text{ S cm}^{-1}$. This achievement places our work at the forefront of conductivity enhancement for cotton-based materials when compared to previous studies. Specifically, Yang et al. (2019) achieved conductivity of approximately 1.0 S m^{-1} using graphene oxide and a dip-coating and chemical reduction method, while Yun et al. (2017) reported a conductivity of 286 S cm^{-1} through an immersion process with gold/graphene. Maneval et al. (2021) demonstrated a conductivity of 1.1 S cm^{-1} with graphene using a dip-and-dry technique. Our results represent a significant advancement in the field of conductive cotton thread fabrication, emphasizing the potential for various practical applications, see Table 6.

Table 6. Comparative analysis of electrical conductivity values: our study vs. previous research.

Substrate	Material	Method	Maximum Electrical Conductivity	Ref.
Cotton thread	Graphene oxide	Dip-coating and chemical reduction	$\sim 1.0 \text{ S m}^{-1}$	[35]
Cotton thread	Gold/graphene	Immersion	286 S cm^{-1}	[36]
Cotton thread	Graphene	Dip-and-dry	1.1 S cm^{-1}	[14]
Cotton thread	Graphene	Dip-and-dry	$2505.675 \text{ S cm}^{-1}$	This work

Third, the effect of temperature on the electrical conductivity of the graphene-based conductive thread was investigated by placing the prepared thread in the oven and changing the temperature from 30°C to 130°C . This study was performed for four conductive threads (see Table 7). The main purpose of this investigation was to study the electrical behavior of the conductive thread under the influence of temperature: is it semiconductor behavior, metallic behavior, or both? As Figure 9 shows, the four conductive threads show the same trend where electrical conductivity decreases with increasing temperature, indicating metallic behavior. The changes in electrical conductivity are from 68.26 to 23.79 S cm^{-1} (44.47 S cm^{-1}) for thread I, from 40.87 to 26.16 S cm^{-1} (14.71 S cm^{-1}) for thread II, from 69.09 to 33.14 S cm^{-1} (35.95 S cm^{-1}) for thread III, and from 118.39 to 45.08 S cm^{-1} (73.31 S cm^{-1}) for thread IV.

for thread 4. These results show that the electrical conductivities of threads I, II, III, and IV decrease by 65.15, 35.99, 52.03, and 61.92%, respectively.

Table 7. The threads used in the investigation of temperature in the electrical behavior.

Thread	Graphene Dispersion in	V (mL)	Annealing Temperature
I	DI	2.5	100 °C
II	DMF	2.5	100 °C
III	DMSO	2	100 °C
IV	Ethanol	2	76 °C

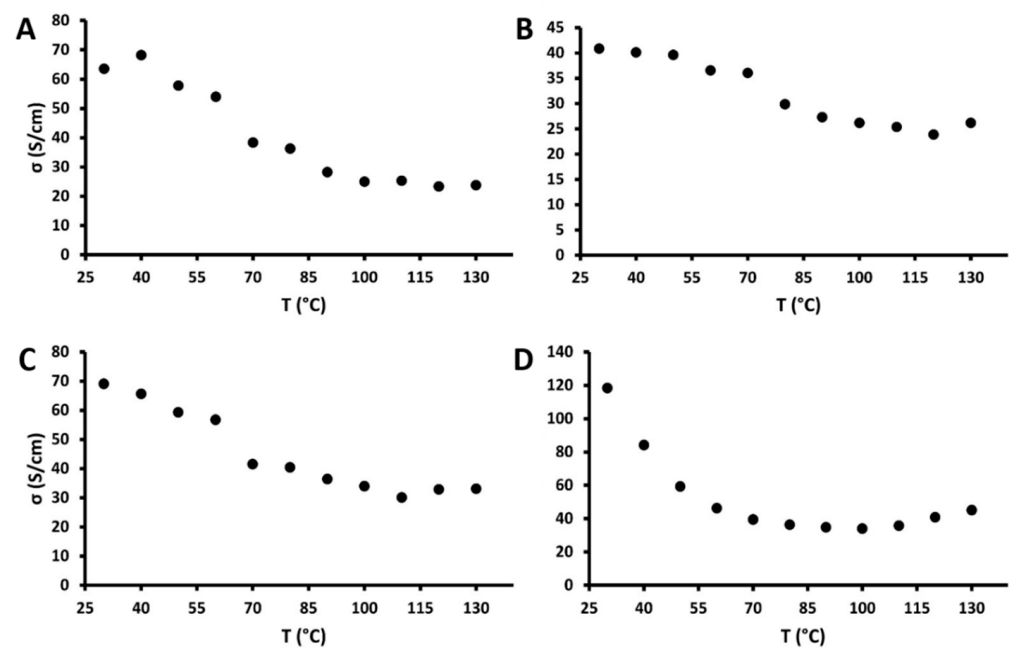


Figure 9. Electrical conductivity as a function of temperature for the conductive threads prepared with graphene dispersed in DI (A), DMF (B), DMSO (C), and ethanol (D).

The decrease in electrical conductivity with increasing temperature can be explained by the following factors:

1. **Thermal Expansion of Cotton Fibers:** Cotton fibers themselves undergo thermal expansion as the temperature rises. This expansion leads to increased spacing between individual cotton fibers in the thread. As the distance between conductive pathways (graphene-coated cotton fibers) increases, it becomes more difficult for electrons to traverse the larger gaps, resulting in higher electrical resistance and reduced conductivity.
2. **Graphene–Cotton Interaction:** The interaction between graphene and cotton fibers can be temperature-sensitive. At higher temperatures, the adhesive or binding properties of the graphene coating on the cotton fibers may weaken or become less effective. This can result in the detachment or reorientation of graphene particles on the cotton surface, leading to interruptions in the conductive pathways and a decrease in electrical conductivity.
3. **Thermal Vibrations of Cotton:** Elevated temperatures cause increased thermal vibrations of the cotton fibers. These vibrations can disrupt the alignment and orderliness of the graphene-coated cotton fibers, leading to a less organized and less conductive structure.
4. **Cotton Degradation:** Cotton fibers may undergo thermal degradation or decomposition at elevated temperatures. This can lead to changes in the cotton's physical and

chemical properties, potentially affecting the conductivity of the graphene-coated cotton thread.

In summary, the decrease in electrical conductivity of the cotton thread treated with graphene at higher temperatures can be attributed to factors such as thermal expansion of cotton fibers, changes in the graphene–cotton interaction, thermal vibrations of cotton, and potential cotton degradation. These factors collectively contribute to the observed reduction in conductivity as temperature increases.

3.6. Washability Study

The main purpose of the washability study is to investigate whether or not the electrical conductivity of the conductive thread changes after several washing and drying cycles. This study was performed for the conductive thread prepared with graphene dispersion in DMSO (1.5 mL). First, the electrical conductivity of the conductive thread was measured before washing and found to be 69.09 S cm^{-1} . The conductive thread was then washed by hand at room temperature for 10 min without using detergent. Before the next wash, the conductive thread was dried at room temperature for 30 min. The electrical conductivity was measured after each wash cycle. As Figure 10A shows, the electrical conductivity decreases as the duration of the wash cycle increases, which is due to the loss of graphene. The results show that the electrical conductivity decreases by 18.64% after the first wash and dry cycle. After five consecutive wash and dry cycles, the electrical conductivity decreased by about 95%, and then a slight change occurred in the electrical conductivity, which reached the minimum value of 0.29 S cm^{-1} after the fifteenth wash and dry cycle. According to the washability results, we need to improve the manufacturing process to maintain and fix the graphene in the thread, but our conductive threads are more suitable for electrical devices that do not need to be washed.

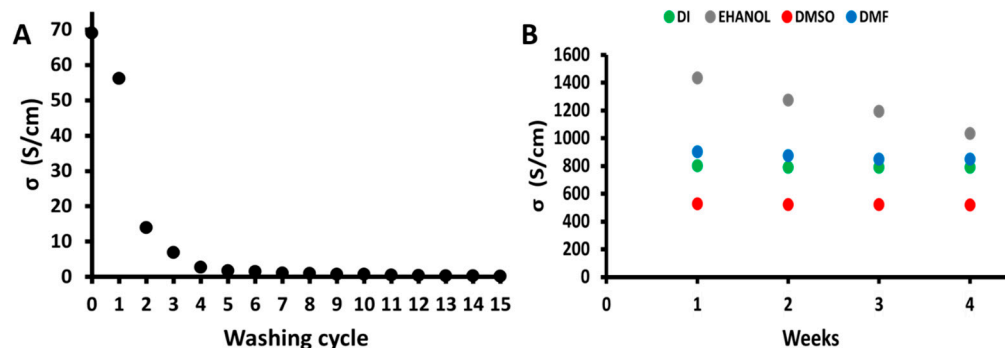


Figure 10. (A) The change in electrical conductivity of the conductive thread as a function of the number of wash and dry cycles; (B) Electrical stability of four conductive threads over a period of four weeks.

3.7. Electrical Stability Study

The electrical stability of conductive threads is the ability of the threads to maintain their electrical conductivity over several weeks. In this study, the electrical stability of four conductive threads was investigated over a period of four weeks, as Figure 10B and Table 8 show. The results showed that the electrical conductivity of thread I decreased by 1.20% from 801.00 to 791.35 S cm^{-1} after two weeks and then remained constant when measured after three and four weeks, indicating that the sample is stable. The electrical conductivity of thread II decreased by 2.94% from 801.00 to 791.35 S cm^{-1} after two weeks, decreased by 3.03% after three weeks, and remained constant when measured after four weeks. Thread III showed more stability, with electrical conductivity decreasing by 0.90% from 527.57 to 523.00 S cm^{-1} after two weeks and remaining constant during the rest of the time. In contrast, the thread IIIV was far less stable than the others threads, with electrical conductivity decreasing by 11.11% from 1433.12 to 1273.88 S cm^{-1} after two weeks and further decreasing by 6.25% and 13.33% after three and four weeks, respectively. From

the study of electrical stability, we conclude that the conductive threads prepared with graphene dispersed on DI and DMSO are more stable than others.

Table 8. Electrical stability study of the conductive threads over a period of four weeks.

Thread	Graphene Dispersion in	Weeks	1	2	3	4
I	DI	Electrical conductivity (S cm^{-1})	801.00	791.35	791.35	791.35
II	DMF		902.33	875.79	849.25	849.25
III	DMSO		527.75	523.00	523.00	523.00
IIIV	Ethanol		1433.12	1273.88	1194.26	1035.03

3.8. Conductive Threads: New Generation

The development of highly conductive thread-based graphene represents a significant advancement in the integration of graphene, cotton thread, and electronic technology, enabling the creation of a novel generation of flexible, cost-effective, environmentally friendly, and lightweight multifunctional threads with electronic capabilities. In this study, we have successfully replaced the conventional power cable of a mobile charger with a conductive thread composed of graphene dispersed in DMF with a measured electrical conductivity of $\sigma = 2416.32 \text{ S cm}^{-1}$. Notably, our observations demonstrate that the charger operates with exceptional efficiency while retaining its flexibility, as Figure 11A depicts and the accompanying supplemental video. Furthermore, we have also replaced the power line within a computer mouse cable with a conductive thread incorporating graphene dispersed in ethanol ($\sigma = 2505.68 \text{ S cm}^{-1}$), and our observations reveal that the device operates with remarkable efficiency, as Figure 11B illustrates and the supplemental video. These findings conclusively establish that our conductive threads represent a groundbreaking advancement in electronic applications, owing to their notable attributes including high electrical conductivity, flexibility, lightweight nature, and cost-effectiveness.

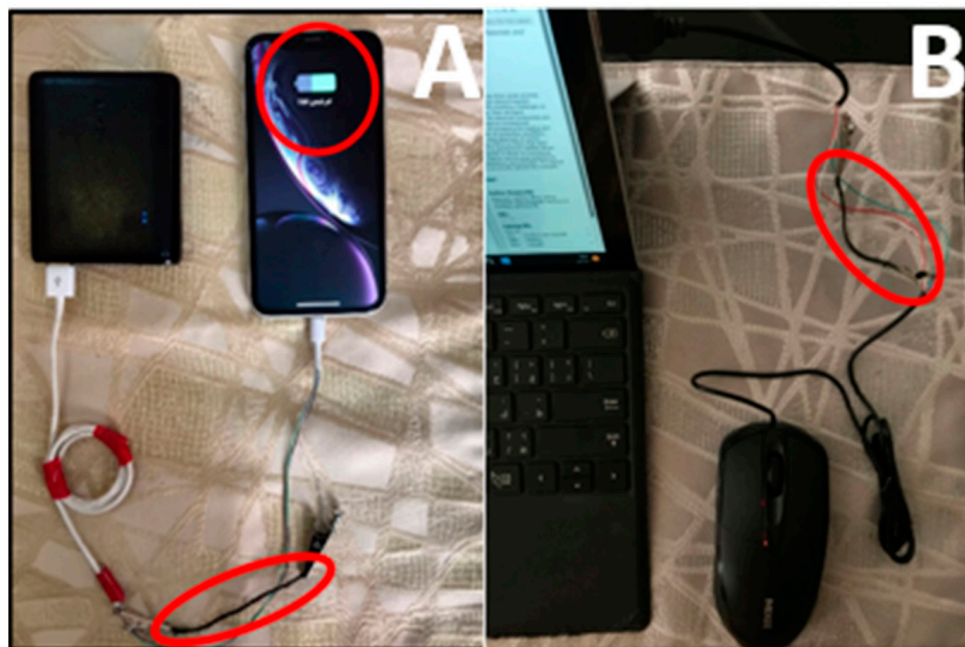


Figure 11. Conductive threads replacing conductive wires in a cell phone charger (A) and a computer mouse (B). The two devices operated with high efficiency.

4. Conclusions

In summary, we have successfully developed a novel approach for fabricating highly electrically conductive cotton threads by incorporating graphene into various solvents,

including deionized water, DMF, DMSO, and ethanol, as a replacement for conventional conductive wires. This fabrication method offers several advantages, including simplicity, eco-friendliness, low cost, and electrical stability. Our investigation revealed that the electrical conductivity of the produced threads is influenced by four key factors: the quantity of graphene dispersion, the annealing temperature, the type of solvent used, and the temperature. Notably, we observed that the maximum conductivity of the conductive thread was achieved when using a high amount of graphene dispersion (6 mL), which can be attributed to the increased concentration of graphene within the thread. Furthermore, the annealing temperature exhibited a significant impact on the conductivity, with variations depending on the specific solvent employed. Remarkably, the conductive thread prepared with graphene dispersed in ethanol exhibited an exceptional electrical conductivity of $2505.675 \text{ S cm}^{-1}$ when annealed at 78°C . These findings highlight the potential of our approach in producing highly conductive cotton threads for various applications. The investigation of electrical conductivity in relation to temperature revealed that all conductive threads displayed metallic behavior. To further characterize the prepared conductive threads, SEM and FTIR analyses were conducted, confirming the presence of graphene. Additionally, elemental analysis using EDS provided further evidence of graphene formation. TGA analysis demonstrated that the thermal stability of the conductive threads improved, with reduced mass loss compared to untreated threads. Based on our research findings, the synthesis of graphene-infused cotton threads holds promise for future applications in the field of electronics, serving as a potential substitute for flexible and conductive wires.

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